SYSTEMS OF ORDER

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In an ordinary solid solution, the different species of atoms are arranged at random on the atomic positions of the lattices. At a given composition AB, any individual lattice point is occupied indifferently by either A or B atoms. Until comparatively recent times this purely random affair was thought to apply in all instances; no order in the atomic arrangement was surmised. However, in late years, it has been determined, largely through X-ray diffraction, that in a multitude of cases the atoms are arranged in as definite a way as are the two different elements of an ionic salt.

Strangely enough, the earliest prediction of ordered structure was not based on X-ray evidence, but on chemical experiments, by Tamman (322, 323). He found that suitably treated Cu-Au alloys of greater than 50 atomic % Au were resistant to HNO_3. With less Au, the Cu atoms of the solution were dissolved but the Au atoms were not. On the basis of this evidence he concluded that a 50 atomic % Cu-Au alloy contained an ordered arrangement of atoms, and any extra Cu atoms did not fit in the organization so were readily removed.

Today, the final evidence of the existence of ordered arrangements, or superlattices, is furnished by the presence of "superstructure lines" on X-ray diffraction patterns. The earliest superlattice lines were observed by Bain (Ref. 23A of 239) for Cu_3Au and by Phragmen (Ref. 25B of 239) for Fe_3Si. Johansson and Linde (Ref. 25A of 239) are responsible for the first analysis of an ordered structure from its X-ray pattern: they worked with the alloy CuAu.

Superstructures are characterized by the feature that in them atoms of one species tend to surround themselves by atoms of the other type. It is well to bear in mind that, while in some cases the atoms surround themselves

as completely as is possible with unlike neighbors, in others they stop definitely short of this limit.

Superlattices form at relatively low temperatures. As the temperature increases, the first effect is only a greater amplitude of thermal vibration of the atoms about their equilibrium positions. When this effect becomes large enough, occasional pairs or small groups of atoms acquire sufficient energy to break away from their places in the lattice and interchange positions with each other. At all temperatures above a certain critical value the usual randomness persists. As the temperature is lowered through the critical point, order sets in, increasing as the temperature drops, and approaching perfection only at low temperature.

Suppose we form an alloy by introducing into a pure metal atoms of a different metal. If the atoms of both types are then allowed to adjust their positions and crystal structure until stable, thermodynamical equilibrium is obtained. The free energy of the system reduces to its lowest value.

Atoms are said to attract one another if the internal energy of the crystal becomes lowered when the atoms are redistributed among the atomic sites in such a way as to increase the number of dissimilar atoms that form nearest neighbors. If dissimilar atoms attract each other to about the same extent as similar atoms, then each atom is relatively indifferent to the type of neighbors it possesses. The two classes of atoms behave as if they were similar and the mixture becomes homogeneous down to atomic properties. A random solid solution is the result. If dissimilar atoms are attracted more than similar atoms, then the tendency is to adopt a structure in which atoms of one kind are surrounded by atoms of the other

kind, in order to obtain a maximum number of dissimilar atoms as nearest neighbors. In true metals, with similar electrochemical characteristics, the resulting structure is usually an ordered solid solution or superlattice.

Order can be subdivided into two categories. It can be described from a local point of view, with reference to the tendency of atoms to be surrounded by unlike neighbors. This phenomenon is referred to as short range order. If this inclination extends for many interatomic distances in a lattice, atoms of one kind will be segregated on one set of atomic positions, leaving atoms of the other kind to the remaining positions. The resultant array can be described as a lattice of A atoms interpenetrating a lattice of B atoms. This segregation of atoms to particular sites takes place with little or no lattice deformation, but gives an ordered solid solution -- a superlattice displaying long range order.

It is evident that a perfectly ordered solution can only occur when the ratio of the numbers of different atoms in the alloy is a simple one, since the alternation of the dissimilar atoms through the structure demands an exact and simple ratio. Thus, today's well founded superlattices are observed in either a 3:1 or 1:1 ratio. In systems showing superstructures at such ratios, partial or imperfect order is often obtained at compositions which differ slightly from the exact ratios. A superlattice can, of course, occur only when the alloy system forms a solid solution extending over a range of composition which includes at least one of these ratios, although fulfillment of this condition in no way assures formation of a superlattice at the critical composition.

The formation of a superlattice may be regarded as a process in which the lattice strain produced by the solute element in the random solid solution is reduced by the formation of an ordered structure. Provided, then, that the solvent and solute atoms are of sufficiently similar size to permit the formation of a wide solid solution, the tendency to form superlattices increases with increasing difference in atomic diameters, since the greater this difference, the greater the strain to be relieved. Apart from this simple effect of atomic diameter, the formation of a superlattice is favored by increasing electrochemical factor. It is probable, therefore, that there is a continuous range, from superlattices involving only the metallic bond and formed merely to relieve the lattice strain resulting from atoms of unequal size, to superlattices with increasing degree of ionic bonding.

Theoretical treatments of superlattices have been made on a basis of investigation. The first were on the concept of lattice using formal thermodynamic relations as a basis. More recent ones used simple assumptions about atomic forces and calculated quantitative results which compared with experiment quite favorably. Any full analysis of this work would be quite extensive, so brief mention of the most important theories only will be made.

Bragg and Williams used as the basic concept of their theory, long range order. They assumed that V, the energy to effect an interchange of a B atom and an A atom, is proportional to the degree of long range order S, according to the relation $V = V_0 S$, where V_0 is a constant representing the interchange energy when order is complete. This assumption leads to a theory predicting a critical temperature corres-

times with, sometimes without, a latent heat. The absence of short range order from this theory is a defect, since it seems certain that the principal interactions in crystals are between very close neighbors.

A logical development of quasi-lattice theory is therefore to consider a concept of order that considers only nearest neighbors, and to compute ordering energies based on nearest neighbor interactions. Theories based on this point of view have been advanced by Bethe and extended by several others, especially Fowler. As long distance order is defined in terms of the number of right atoms, so short range order is defined in terms of the number of right pairs (right pair being one of unlike atoms -- as AB pairs).

With the foregoing outlined summary of the order-disorder concept and its phenomena, let us proceed to compile a list of alloys which various authors have found to exhibit the formation of a superlattice. The data presented is the result of a literature survey by this author covering from 1938 through the present date. Quite frequently, the paper from which the information was compiled gives but very skeleton data on the reaction. Such systems are generally mentioned only as a side issue in a paper on some subject quite remote from the principal topic of order-disorder reactions. Similarly, some systems listed have been reported as forming a superlattice by a single author, with no confirmation by others. The scope of the present paper is not to attempt to justify further investigation on alloys where complete data is lacking. Rather, the author will merely record such material which is here available without

comment as to its scientific authenticity.

The systems will be compiled in alphabetical order, using the component with highest alphabetical sequence of recognized English scientific name (not symbol), as a class of reaction present in the system.

Systems Said to Order

Cr-Al

A superstructure has been reported (Ref. BMC p. 239) for tetragonal Cr_2Al. This structure forms upon the body-centered cubic lattice. If we consider planes of the (100) type, which are formed alternately of cube corners and of cube centers, we find that every third plane contains Al atoms and the remaining planes Cr atoms.

Al-Co-Fe

A superlattice structure was found (72) in the β phase for alloys with less than 50 atomic % Al annealed from 800° C.

Ni-Co-Al

Equilibria have been shown (21) in quaternary diagrams for Ni-Co ratios of 75:10, 80:20, 50:10, and 20:30. In this region there are four phases, of which the γ' is FCC, ordered, and β-β' is BCC, ordered. The γ' phase in the Ni-Al system at about 13% Al extends to about 2% Co at 1000°C and 3% Co at 200° C in the ternary system.

Al-Cu

A superstructure formed prior to the θ' phase in the Al-Cu. Cu-Al alloy is described (13). This indicates a system of three tetragonal structures oriented with basal planes parallel with the (100) planes. It is suggested that an examination of the Cu-Al (001) planes

has occurred.

Cu-Mn-Al

The Heusler alloy, Cu_2MnAl in the ferromagnetic condition is highly ordered (Ref. 3A of 239). The ordered state results from a rapid cooling from a 600C annealing temperature. The superstructure atoms are distributed in a BCC cell.

Fe-Al

As Al is added to the BCC Fe lattice, the Al atoms replace Fe atoms at random until a composition of 18% Al is reached. Here, the solute Al atoms cause a diffuseness (Ref. 3A of 239) attributed to the onset of order in the annealed alloys. At 25% Al, slow cooling produces the ordered Fe_3Al lattice in which not only the nearest neighbor of an atom, but also its next nearest neighbors are unlike atoms. In the Fe_3Al lattice, Al atoms take such positions that, in following the BCC lattice along a cube diagonal, one meets Al atoms at alternate small cube (BCC) centers. On addition of further Al, the solute Al atoms begin to fill in the centers of the additional small cubes. At Fe Al, both rapid cooled and annealed alloys require an ordered structure of the C. Cl type, with cube centers occupied by Al atoms and the corners by Fe atoms.

Fe-Ni-Al

There is microscopic evidence of ordered and disordered phases coexisting in FCC Alloys (41). In alloys of Fe, Ni (18-30%), and Al (9-17%) quenched from 11[illegible] - 12[illegible] C, and annealed at 600C decomposes to $\alpha + \alpha'$, followed by an ordering of α' (74). If the alloy

is cooled from above 1100° C at a controlled rate decomposition of α into $\alpha + \alpha' + \gamma$ occurs at 7[illegible]0° C; below this temperature ordering of α' occurs.

Ni - Al

In an alloy of 31.6 atomic % Al, the Beta phase undergoes partial disordering at high temperature and becomes duplex. The ordered structure is of the cubic Cs Cl type (1[illegible]).

Al - Ag

The first stage (131) in precipitation in Ag-Al alloys is the collection of Ag atoms in nuclei with partial order in Al atoms.

Cu - Sb

Alloys with Sb - 64.0at% Cu consist of homogeneous Beta phase, a cubic structure with 16 atoms per unit cell; lattice parameter is 5.914 A. An orderly arrangement (3) permitting the Fe Al_3 structure is described. It corresponds to the formula $Cu_{12}Sb_4$ (Mg_3Sb). Ordered arrangements of Mn_3Sb and Mn_2Sb have been reported, but the author has no specific information on these structures.

Ni - Sb

The formation of a superlattice from the α phase is confirmed (288). The ordered structure α' ($Ni_{4.5}Sb$) appears at 1400 C; its range of existence at room temperature is 88 - 90 atomic % Ni. The alloy is paramagnetic.

Sb - Ag

A range of alloys, 1[illegible]-31% Sb, is investigated (3.4). Two intermediate phases are confirmed, ε, a HCP lattice, and a disordered

phase, roughly Ag_3Sb which exists at both high and low temperature modifications. The transition is correlated at [illegible]° - [illegible]° C, according on composition, and is interpreted as an order-disorder change.

Cu - Be

The disordered crystal is BCC; the ordered structure is a cubic of the Cs Cl type.

Ag - Cd

As above, the disordered crystal is BCC; the ordered crystal is of the cubic Cs Cl type.

Mg - Cd

Below 2[illegible]° C, superstructures appear at the compositions Cd_3Mg, Cd Mg, and Cd Mg_3 (2). Superlattices have been found with Cd ranges from 7[illegible].2% to 93.[illegible] (147). Both the Mg_3Cd and $MgCd_3$ ordered structures possess the CPH lattice.

Mg - Ag - Cd

The quasibinary system, Mg-Ag_3Cd_3, exhibits a CPH ordered structure in the range 1[illegible]-70 atomic % Mg.

Mg - Cd - Zn

A superlattice has been reported (147) at the composition 72% Cd and 1[illegible] Zn.

Ni - Cr

Electrical and dilatometric measurements in the Ni-rich Cr-Ni solid solution exhibit an anomaly correlated to be due to an order-disorder transformation similar to that in the Cu-Zn -Ni solid solution (365).

Cr - Pt

A superstructure of what is thought to be Cr_3Pt has been reported (107). Another Author (118) finds a superstructure in alloys of 37-61.6 atomic % Cr on slow cooling from 1330° C.

Fe - Co

These alloys, in the composition range 40-65 % Co, order from a BCC to a cubic of the Cs Cl type. T_c = 732° C (91). Ordered alloys exhibit a larger lattice constant at room temperature than quenched, disordered material, owing to the lower expansivity of the ordered alloy (96).

Co - Pt

One investigator (154) finds that a cast sample of Co Pt contains a mixed structure, FCC (a = 3.75), and BCC (a = 2.85) similar to the Cs Cl cell. A quench from 1300° C results in FCC structure alone. A volume increase of 14% when BCC forms from FCC is the author's basis for concluding that the BCC must become ordered. More recent papers are probably more accurate. There is general agreement that the disordered lattice is a FCC with a_o = 3.741 - 3.744, and the ordered structure is an Au Cu -- like FCT lattice with a_o = 3.785 - 3.818 and C = 3.639 - 3.668 (109,153). T_c = 820°C. This work relates to a 50 - 60 atomic % composition.

Au - Cu

It is on this best known of all ordering systems that most work has been published. Disordered alloys of this system possess a FCC structure. In the Cu Au range, a superlattice forms in which alternate layers of Cu and Au atoms form an (001) plane on the FCC

solid Solution lattice, distorting it into a tetragonal structure. T_c for this reaction is about 375° C (117). Between 370° and 400° C, another structure, referred to as Cu Au II, is stable. It is an orthorhombic lattice, with two nearly equal axes, and the third about ten times as long (182). At Au Cu_3 is found another ordering process, FCC disordered to FCC ordered (25[illegible]). Ordered Au Cu and Au Cu_3 are related by an intermediate series of phases (289). At 75 atomic % Cu there exist indications of long range order at 196° C (116). At 370° C 0.[illegible]% of the Au Cu lattice, [illegible]% of the Au Cu_3, and 4% of the Au_2 Cu_3 are not in ordered positions (3[illegible]). The critical points of these alloys are raised by pressure (3[illegible]).

Au-Cu-Ni

High concentrations of Ni do not affect transformations of the Au-Cu system. The effect of Ni on Au Cu_3 ordering is less than with the Au Cu lattice (261).

Au-Cu-Ag

Addition of Ag lowers the critical temperature of the Au-Cu system (165,164). Addition of 5% ag inhibits ordering completely (51).

Cu-Mn-I

The ternary phase Au Mn I shows a complex order-disorder change resembling somewhat that in Au Cu_3, but involving ordered and disordered arrangements of both metal atoms and vacant lattice points.

Cu-Ni-Mn

Recrystallization in a [illegible] Cu [illegible] Ni, 2[illegible] Mn alloy is considered to be due to an ordering process, from a cubic to a [illegible] lattice (279)

Cu-Mn-Sn

With greater than 20 atomic % Mn, the Beta phase has an ordered structure which, for a Cu_2MnSn alloy, persists at 630° C (54).

Cu-Pd

Two structures in this system have been found to exhibit ordering. Cu_3Pd and $CuPt$. Cu_3Pd orders to a cubic of the Cs Cl type from the disordered BCC lattice, except in the range 37-42 atomic % Pd, in which the Cs Cl cubic forms from a disordered FCC. Pd_3Cu orders from an FCC to an ordered FCC. The latter is slightly distorted from the cubic, investing it with tetragonal symmetry (67).

Cu-Pt

In this system near the 50 atomic % composition, the FCC disordered lattice becomes striped (herringbone) with alternate planes of Cu and of Pt atoms on (111) planes. This structure has tetragonal symmetry (Ref. 27 of 239,268). A 44 atomic % Pt (268) and Cu_3Pt have been found to agree with this concept. Order in the composition range 5-38 atomic % Pt exists only below 450°C, with the maximum critical temperature at 20 atomic % Pt (93). The lowest degree of disorder exists in alloys containing a slight excess of Cu over the Cu_3Pt composition. Alloys between 37 and 42 atomic % Pt change from a disordered FCC to the Beta brass type structure, ordered BCC. $PtCu_3$ changes from a disordered to ordered FCC lattice. One author writes (93) that ordered mixed phases exist between 30 and 90 atomic % Pt, while another (268) believes no order exists above a composition of 60% Pt. Degree of disorder (93) here has as follows: $CuPt$, $CuPt_3$, $CuPt_7$.

Cu-Zn

In alpha brass, short range order is persistent up to the melting point (144). The alpha phase lattice is ordered being similar to the 50 at Cu Cu alloy (FCC to ordered simple tetragonal). At room temperature, alpha phase exists a long time, changing to a cubic alpha on tempering (144). Disordered beta brass is a random BCC lattice. On ordering, a Cs Cl type BCC is produced in which a_0 = 2.9 2 A (11). Tc is about [illegible] C.

Au-Ni

This system forms a BCC of the Cs Cl type on ordering from a disordered BCC.

Au-Pb

The alloy Au Pb_2 is highly ordered near the fusion temperature (199).

Au-Sn

This is another system exhibiting a high degree of order near the melting point (197).

Au-Zn

The beta phase, with limits of homogeneity of 47.5 to 51.0 at % Au, has Cs Cl type structure of 2 atoms per unit cell, with maximum order at equal atomic percentages of Au and Zn (1). The order-disorder reaction sets in on heating above 60.0 C, but does not proceed to completion up to the melting point of 725° C.

Ag-Hg-I

The Ag_2 Hg I ternary phase shows a complex order-disorder change resembling somewhat those in Au Cu3.

Fe-Ni

Experimental evidence indicates a superstructure in the neighborhood of Ni_3Fe (153). Super-lattice lines at 74.3 atomic % Ni have been found (208). T_c is around 500° C, but the transformation is remarkably sluggish. The reaction is probably of the discontinuous type (181). The disordered lattice is FCC, as is the ordered product.

Mn-Fe-Ni

The alloy (Mn_2FeNi_2) exhibits an ordering reaction in changing from a random FCC to an ordered FCC.

Fe - Pd

On annealing below 640° C, Fe Pd forms an ordered tetragonal with an axial ratio of less than unity. The superlattice is most stable with an excess of Pd. At 60 atomic % Pd, it is stable up to 775° C, while stable only to about 675° C at 51.9% Pd. There exists also an $FePd_3$ ordered state with the same structure (FCC) as $AuCu_3$ (164).

Fe-Pt

The disordered Fe Pt exists as a FCC and at about 500° C (316), orders to the Au Cu type tetragonal face centered structure.

Fe-Pt-Rh

An alloy of 50 atomic % Fe, 45 atomic % Pt and 5 atomic % Rh orders in the tetragonal structure of the Cu Au type (Ref. 360 and 239).

Fe-Rh

At 5 atomic % Rh, this alloy forms an ordered structure (235).

Fe-Si

Some magnetic measurements (Ref. 3[illegible] of 239) indicate ordered phases at 12.5 and 25 atomic % Si. X-ray evidence (Ref. 2[illegible] of 239) establishes only the latter, the Fe_3Si ordered phase, which is a BCC of the Cs Cl type.

Ag-Mg

The beta phase orders (transforms to the alpha phase (FCC)) but ordering is most complete in the region of 50 atomic % Mg. There is some evidence, but no full investigation, for a superlattice in the alpha phase in the 25 atomic % Mg region. The beta phase orders from a BCC to a BCC of the Cs Cl type.

Ni-Mn

The composition $Mn\ Ni_3$ orders from a random FCC to an ordered FCC. The transition temperature has not been agreed upon by investigators. Estimates include 710° C (326), 520° C (337), less than 950° C (33[illegible]), and 3[illegible]0-400° C (185) with the remark that the transition occurs over a range, no real critical temperature existing. One author reports the formation of a FCT with axes at 1[illegible]00 C (331).

Pd-Mn

A superlattice, Pd_3Mn, at 1[illegible] Mn has been reported (137). The reaction steps observed were: FCC --> 1165° C --> tetragonal --> 530° C --> ordered tetragonal.

Sn-Mn

Superstructures have been reported for compositions of $Sn\ Mn_2$, $Sn\ Mn_3$, and $Sn\ Mn_4$. No amplifying data were found.

Ni-Pt

A tetragonal, ordered Ni Pt phase was observed (92), with 1000 C limits of 40-55 atomic % Pt.

Ni-Sn

At 950° C occurs an order-disorder transformation in the beta phase. Ni_3Sn is the superlattice; it is a CCH of the Mg_3Cd type.

Ag-Pt

In Cu region 25-35 atomic % Pt there is a new phase, Ag Pt, which at less than 800° C has a simple cubic structure; at greater than 800° C it becomes disordered (263).

Ag-Zn

In the range 25-35 % Zn is a change of disordered to ordered lattice (Beta Beta one) taking place at 230-225° C. Similar to the Beta brass structure, this alloy exhibits the ordered BCC of the Cs Cl type.

As a final project, let us look briefly to the effect of the order-disorder phenomenon on various properties of an alloy.

Since the ordered state is the one of lowest energy, energy must be supplied to create disorder. During disorder, the specific heat is greater than normal; the value at each temperature depends on the rate of change of order with temperature. A latent heat is displayed by some systems, not by others. Theory is weak in predicting this circumstance.

Electrical resistivity is affected by ordering. If the disordered state is retained at a temperature below the critical by a quench, the strains from the distribution irregularities are superimposed upon the disturbances from thermal agitation; each contributes

to electrical resistance. Annealing induces order, lowering the resistivity.

Magnetic properties are order-dependent. The Heusler alloys become ferromagnetic in the ordered state. Diamagnetic susceptibility increases for some alloys (e.g., Au Cu$_3$, Cu Pd) upon ordering while it decreases in others (e.g., Au Cu).

Whether of the long or short-range type, ordering is always accompanied by a diminution of volume.

Young's Modulus increases with ordering in Cu$_3$ Pt and Au Cu$_3$ but decreases in Au Cu and Cu Ni. Plastic constants are sensitive also to local order.

Mechanical properties are also altered when ordering occurs. Hardness, tensile strength, and the elastic limit are generally increased.

Plastic deformation tends to destroy long-range order.

Any major attempting a complete analysis of the subject, order-disorder, would of necessity fill volumes. Suffice it to say that knowledge and theory of the phenomenon is at least in its adolescence stage. Modern work is, however, making rapid strides towards correlation of theory and experiment, and gives promise of supplying in a few years a workable knowledge of the order-disorder concept.

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